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Feasibility of foam forming technology for producing wood plastic composites

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Abstract

Cellulose fiber-containing thermoplastic composite materials are being used in an increasing number of applications produced typically by injection molding and extrusion processing methods. One potential way to manufacture thermoplastic cellulosic fiber composites is foam forming technology developed originally for paper manufacturing. This article compares the low-density polyethylene (LDPE) and unrefined northern bleached softwood kraft pulp (NBSKP) composite materials prepared with foam forming, extrusion, and injection molding. The results show that the foam forming enabled three times higher Charpy impact strength properties and 68% higher tensile modulus compared to injection molded 30% NBSKP fiber-containing LDPE composites without changes in composite color. Foam forming is a potential large-scale manufacturing method for thermoplastic composite sheets used, for example, in compression molding or thermoforming.

KEYWORDS

cellulose, composites, manufacturing, molding

1 | INTRODUCTION

Thermoplastic composites with ligno-cellulosic fibers from short cellulose to long hemp and flax type fibers are finding their way into an increasing number of applications.[1] The positive advantages of natural fibers and cellulose compared to the more commonly used glass fiber reinforcement are good availability, originating from a renewable resources, low weight, low cost, relatively good mechanical properties such as tensile modulus and flexural modulus, improved surface finish of molded parts composite, less damage to processing equipment, and minimal health hazards.[2] However, there are challenges in manufacturing of thermoplastic wood or natural fiber composites. The polymer matrix should be selected so that ligno-cellulosic fibers do not degrade during processing due to high temperature. The processing temperature should preferably be below 200°C.[3] The thermoplastics that soften below this temperature are, for example, polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), and polystyrene (PS). By selecting one of these hydrophobic polymers as a matrix, compatibility challenges will occur due to hydrophilic cellulosic fibers. Fiber dispersion and fiber-polymer matrix connection related issues are in many cases improved by using different coupling agents or using physically or chemically modified fibers.[4]

Manufacturing of ligno-cellulosic fiber-containing thermoplastic composites or wood plastic composite (WPC) are typically made using extrusion, injection molding, compression molding, or thermoforming (pressing) processes. For separate short fibers or sawdust,

extrusion and injection molding are common processes.[5,6] In compression molding and thermoforming processes typical for polymer sheets or polymer granules, the addition of natural fibers can be made using needle punched long fiber (e.g., flax) nonwoven-type mats impregnated with polymer and formed into products. Another way is to introduce thermoplastic fibers in the natural fiber structure or to use woven structures combined with thermoplastic to get very strong structures. These mats are thermally formed to shapes needed in end products using molds.[7–11]

One way toward high-performance composites is to utilize the water-forming method, commonly used in paper and nonwoven manufacturing, in the production of ligno-cellulosic structures together with thermoplastic polymer followed by compression molding of material.[12,13] Another large-volume manufacturing method used to some extent in the paper and nonwoven industries is foam forming. However, VTT has extensively developed the foam forming process and foam-formed products also in application areas other than printing papers, packaging boards, and nonwovens. The foam forming technology is seen as a potential manufacturing technology to produce high-performance and cost-efficient thermoformable WPC materials. Foam forming technology utilizes small air bubbles containing aqueous foam as a transporting medium for solids like fibers. Air bubbles effectively prevent fiber flocculation by decreasing mobility of fibers leading to excellent homogeneity of a product.[14,15] Foam also increases the distance between fibers enabling the production of very high porosity structures, which can be used in product weight reduction.[16] The utilization of a wide variety of different fiber materials, from nanofibers to several-centimeter-long fibers as well as particles heavier and lighter than water, is possible in the foam forming process.[17,18] The reinforcement capacity of wood fibers can be maximized, because the fiber length is maintained in the foam forming process like in water-forming. During forming, the consistency can be increased substantially with foam compared to traditional water forming.[15] In the foam forming process solids, water and surfactant are mechanically mixed to generate solids containing foam with an air content between 30 and 70%. After the foam generation phase, the solids containing foam is spread on a wire mesh and the foam is drained with a vacuum. The formed web is wet-pressed and finally the material is dried by contact or noncontact drying methods.

The objective of this study was to compare the structure and material properties of cellulose fiber-containing polyethylene composites produced by foam forming, extrusion, and injection molding. An additional motivation was to see if there is potential for relatively short fiber cellulose composites manufactured by foam forming to enter in the business of thermoplastic composites. The novelty of this study is that WPC produced by foam forming technology does not exist on markets yet.

2 | EXPERIMENTAL

2.1 | Materials

WPC materials were produced from unrefined northern bleached softwood kraft pulp (NBSKP) from Metsä Fiber and low-density polyethylene (LDPE) FA3221 (CAS no. 9002-88-4) from Borealis with MFI 0.3 (190°C/2.16 kg) by three different technologies: foam forming, extrusion, and injection molding. LDPE with MFI value was chosen to ensure proper processing with extrusion and possibility to grind it for suitable form to be added in foam forming. For NBSK pulp, the length-weighted average fiber length and fiber width measured with the L&W STFI Fibermaster fiber analyzer were 2.1 mm and 28.6 μm, respectively. For foam forming trials, the LDPE was ground to powder using a Scheer plastic pulverizer (Reduction Engineering Scheer, Kent, OH). The average particle size of powder measured with a Malvern Instruments particle size analyzer (Malvern Instruments Ltd., Malvern, United Kingdom) was 460 μm and 90% of particles were below 970 μm. Sodium dodecyl sulfate (SDS) (CAS no. 151-21-3) from Sigma-Aldrich was used in foam forming as a surface-active agent to produce foam. Maleic anhydride grafted polyethylene (PE-MAH) (CAS no. 9006-26-2), Licocene PE MA 4351 from Clariant, and a fiber dispersing additive Struktol TPW104 from Struktol Company of America, that is a blend of aliphatic carboxylic acids salts and mono and diamines, were used in plastic processing as a coupling agent and lubricant for cellulose fibers and LDPE.

2.2 | Foam-formed composites

Fiber-polymer foam was prepared by mixing water, SDS, NBSKP, and LDPE powder in a cylindrical vessel (designed by VTT) with an inner diameter of 192 mm (Figure 1). The mixer speed was 3,800 rpm and the diameter of the planar mixing plate was 83 mm. The amount of SDS was 0.6 g/L and the consistency of pulp/LDPE powder suspension was 2.3%. The initial volume of the suspension was constant at 3 L. The foam generation was continued until the air content of the fiber-polymer foam exceeded 60%, corresponding approximately to a 2 min mixing time. The fiber-polymer foam was poured into a hand-sheet mold (designed by VTT) of the size

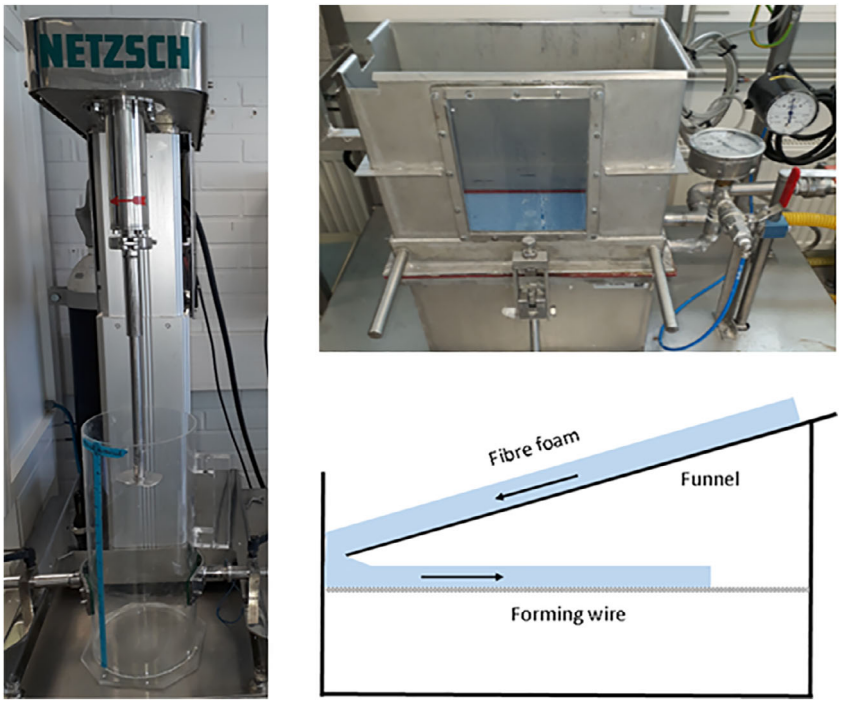

FIGURE 1 On the left, the mixer and mixing vessel for generating fiber-polymer-containing foam. On the right top, the laboratory sheet mold and on the right bottom, the schematic picture of the pouring phase of fiber-polymer foam [Color figure can be viewed at wileyonlinelibrary.com]

350 mm × 220 mm with the help of a funnel (Figure 1). Then, the fiber-polymer foam was spread from one end of the mold to the other and laid on a supported forming wire. The funnel was removed and a plastic cover was placed on the top of the fiber-polymer foam to create a seal for the vacuum used in the forming phase. The foam was removed with a vacuum for about 10–15 s. The formed wet web was removed from the mold and wet-pressed between blotter papers using a pneumatic laboratory sheet press (Lorentzen & Wettre, Kista, Sweden). Wet pressing was carried out in two steps. The pressing level was 7 bar in both steps. At the first step, the pressing time was 5 min and the second step 2 min. The blotter papers were replaced between the pressing steps. Wet-pressed samples were dried between blotter papers by a cylinder drying device (Lorentzen & Wettre). The drying temperature was 70°C and the drying time was 6 hr. Finally, samples were thermo-pressed between two hot plates using a hydraulic laboratory sheet press designed by VTT (Figure 2). Thermo-pressing was carried out in two similar steps, in which process parameters were a temperature of 115°C, pressure of 95 bar and a pressing time of 5 min. Samples were turned over between the thermo-pressing steps. Foam-formed materials were made with 30, 50, and 100% cellulose fiber content.

2.3 | Composites by plastic processing

Before mixing with LDPE the fluffy NBSKP was pretreated with a compactor device, presented in a patent

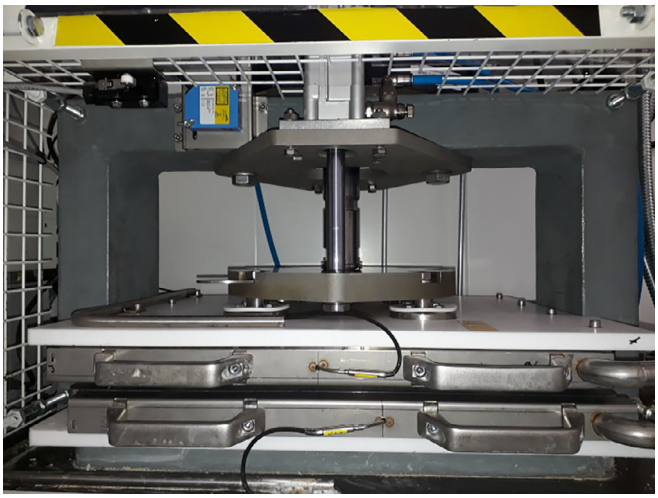

FIGURE 2 Sheet pressing in a hydraulic press [Color figure can be viewed at wileyonlinelibrary.com]

application by Immonen et al.[19] to a form that could be more easily fed into a compounder. The compacted cellulose pellets were dried before compounding to a dry material content of 98.5%. Compounds were made in 30 and 50% cellulose fiber content containing PE-MAH (1%) and Struktol TPW104 (1%) added straight to the compounder with LDPE. In compounding a corotating twin-screw extruder Berstorff ZE 25x33 D (Berstorff GmbH, Hanover, Germany) was used in a temperature range of 60–160–170–180–185–185–190–190–195°C. Diameters and lengths of the screws of the twin-screw extruder were 25 and 870 mm, respectively. The retention time inside the compounder was 1 min.

TABLE 1 Samples prepared for comparison of processes

Sample code	Sample	Thickness (mm)	Density (kg/m^3)
FF-NBSK100	Foam-formed NBSKP 100%	2	0.41
FF-NBSK50	Foam-formed NBSKP 50%, LDPE 50%	2	0.79
FF-NBSK30	Foam-formed NBSKP 30%, LDPE 70%	2	0.93
E-NBSK30	Extruded NBSKP 30%, LDPE 70%	2	1.002
IM-NBSK30	Injection molded NBSKP 30%, LDPE 70%	4	1.038
IM-NBSK50	Injection molded NBSKP 50%, LDPE 50%	4	1.144
IM-LDPE100	Injection molded LDPE 100%	4	0.92

Abbreviation: NBSKP, northern bleached softwood kraft pulp.

Pure LDPE and LDPE-NBSK compounds were injection molded with an Engel ES 200/50 HL injection-molding machine (Engel Maschinenbau Geschellschaft m. b.H, Schwefberg, Austria) and made into dog bone-shaped test bars according to ISO 527-2. The temperature range in injection molding was 180–185–190–190°C, and 200°C in nozzle, the mold temperature was 25°C. LDPE-NBSKP compound with 30% cellulose content was also extruded to 10-cm-wide and 2-mm-thick cast film using Brabender PlastoGraph EC plus (Brabender OHG, Duisburg, Germany) with an extrusion head single-screw Extrusiongraph 19/25D and film calibration unit and temperature range from 180 to 195°C and retention time below 2 min.

Table 1 summarizes all samples prepared by foam forming, extrusion, and injection-molding processes and the materials' thicknesses and densities for comparison.

2.4 | Characterization

2.4.1 | Scanning electron microscopy and light microscopy

In order to see the fiber-polymer morphology in the composite, a scanning electron microscopy (SEM) analysis was made for the cross-section surface using JEOL JSM T100 (JEOL Ltd., Tokyo, Japan). For cross-section the test samples were cooled in liquid nitrogen and broken.

The fiber morphology after processing was also analyzed by light microscopy Leitz Diaplan (Leitz, Wetzlar, Germany) after dissolving out the LDPE matrix with Soxhlet extraction. Pieces of composite samples (2–3 g) were placed into the Soxhlet thimble and fluxed with a hot solution of xylene for 48 hr.

2.4.2 | Particle size analysis

A particle size analysis was run for fibers after dissolving the LDPE matrix out and using Malvern Particle size analyzer (Malvern Instruments Ltd., Malvern, Worcestershire, United Kingdom). LDPE was dissolved from composites with boiling xylene until fibers were clearly separated (32 hr). Particle size analyzer was used instead of fiber analyzer due to high amount small fiber fractions in injection molded and extruded materials compared to foam-formed material.

2.4.3 | Mechanical tests

Tensile strength properties and Charpy impact strength were tested for the materials. In testing, the injection molded specimens were used as such. Samples for tensile strength and Charpy impact strength tests of extruded and foam-formed materials were die cut out of the sheets into the dog bone-shaped specimens with the same length, width, and shape as injection molded samples. Both tests were done using at least five parallel specimens. Tensile tests were performed according to ISO 527 using an Instron 4505 Universal Tensile Tester (Instron Corp., Canton, MA) with a 10 kN load cell and a 5 mm/min cross-head speed. Charpy impact strength for un-notched specimens was tested according to ISO 179 using a Charpy Ceast Resil 5.5 Impact Strength Machine (CEAST S.p.a., Torino, Italy). The test specimens were kept in standard conditions (23°C, 50% relative humidity) for at least 5 days before testing.

3 | RESULTS AND DISCUSSION

3.1 | Visual outlook and morphology

The processed materials and their visual appearance are presented in Figure 3 and exploded view with light behind the sample in Figure 4. In general, injection molding gives freedom of shapes for potential products restricted by mold shape. Extrusion and foam forming are manufacturing methods for sheet- and film-form

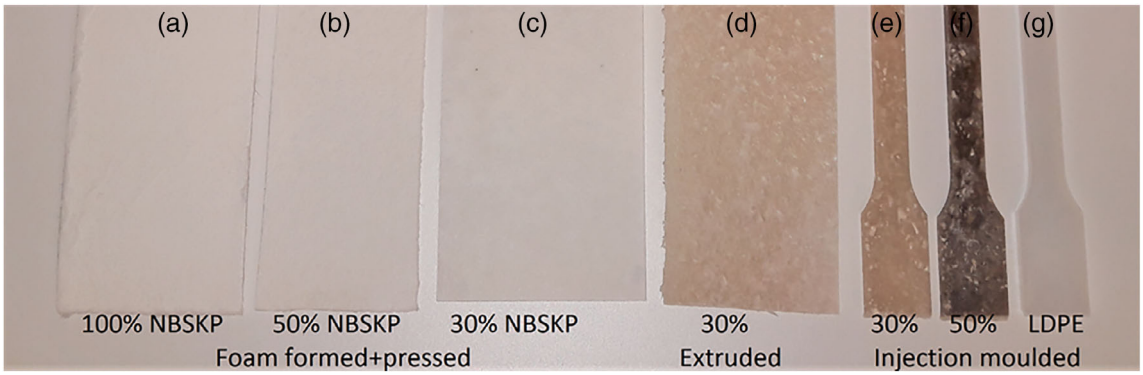

FIGURE 3 Samples from left to right. Foam-formed materials with pure NBSKP fibers and 30 and 50% NBSKP fiber-containing samples (FF-NBSK100, FF-NBSK50, and FF-NBSK30), extruded sheet with 30% fiber (E-NBSK30), and injection molded test bars with 30 and 50% fiber and neat LDPE (IM-NBSK30, IM-NBSK50, and IM-LDPE100). NBSKP, northern bleached softwood kraft pulp [Color figure can be viewed at wileyonlinelibrary.com]

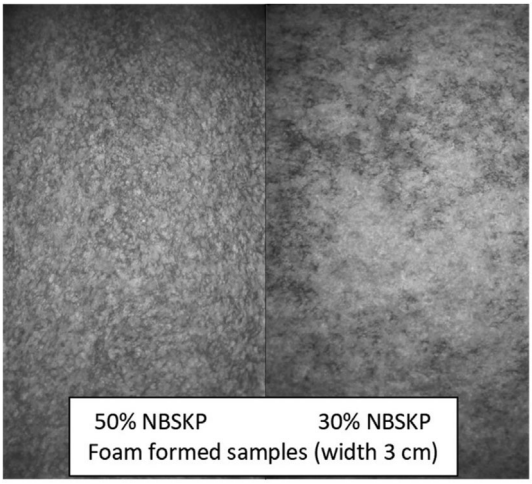

FIGURE 4 Picture of foam-formed and pressed samples with 50% NBSKP (left) and 30% NBSKP (right) in LDPE. Light behind the sample to show fiber distribution in 3 cm wide samples. NBSKP, northern bleached softwood kraft pulp

materials that can be thermoformed to different shapes in the z-direction from an x-y-sheet restricted by elongation of the material at an elevated temperature.

Figure 3 presents the visual difference between materials that were produced with different methods even though they contain similar raw material combinations. In injection molding and extrusion processes the LDPE and cellulose fibers were mixed together in a polymer melt in high temperature, up to 195°C. That is causing color formation in the extruded and injection molded composite materials (d, e, and f), assumingly due to thermal degradation of cellulosic material.[20] The higher the load of the cellulosic material, the darker the product is, which can be seen by comparing samples in Figure 3e,f. The color difference is due to increased melt viscosity of the fiber-filled polymer melt due to higher filler content causing friction and higher shear for the material in the process. The high shear forces are present especially during injection molding, where the mold gate diameter can be small and flow patterns complicated inside the mold. In the extrusion process, the shear forces in the final processing are lower than during injection molding, providing less thermal stress to the cellulosic material and a slightly lighter color for the material presented in Figure 3d (30% cellulose containing extruded material) compared to injection molded material (e) with the same cellulose fiber content. The material prepared using plastic processing contains a significant amount of NBSKP agglomerates, because no special fiber dispersing additives were used on fiber surface to reduce the hydrophilic properties of the NBSKP fiber enabling better fiber-polymer dispersion. This challenge of poor dispersion of cellulose fibers to LDPE without dispersion-enhancing additives is reported also in other studies.[21,22] The lubricant in use was not effective enough to provide proper fiber dispersion.

In foam forming, the temperature is lower during the sheet forming process and even the thermal pressing of the composite sheets can be done at lower temperatures than in typical plastic processing. Foam forming into a sheet is a room temperature process, sheet drying was made in 70°C and thermal pressing in 115°C. Lower processing temperatures and very low or no mechanical shear forces during foam forming saves cellulose fibers from degradation and color formation, which can be seen in Figure 3 (samples a–c). The challenge in foam forming is to achieve even material distribution for hydrophobic and small LDPE particles in an aqueous environment. The critical phases affecting LDPE material distribution are mixing, foam formation and sheet formation. Challenges in mixing and foam formation phases are related

mainly to different surface properties and densities of cellulosic fibers and LDPE and can lead to uneven LDPE material distribution in the planar as well as in the thickness direction of the sheet material. During sheet formation and drying, the water layer prevents effective penetration of hydrophobic LDPE particles between cellulose fibers.[23] After the sheet forming phase, the distribution of LDPE particles in the thickness direction can also be uneven, because of poor adhesion of LDPE particles in fibers combined with one-sided water removal during sheet forming phase. During thermal pressing the LDPE melts, showing a clear difference between the areas containing high or low amounts of LDPE in the sheets. For example, in Figure 4 the uneven distribution of LDPE can be seen even visually as a galvanized or cloudy appearance in high LDPE-containing areas in sample with 30% NBSKP fibers (Figure 4, right sheet). The sample with higher, 50%, fiber amount (Figure 4, left sheet) showed quite even structure and visible network of fibers.

Cross-sections of the composite materials with 30% NBSKP content can be seen in the SEM pictures in Figure 5, where an overview of materials with ×100 enlargement and closer look with ×1,000 enlargement is presented.

In Figure 5a,d are of foam-formed material after pressing. It can be seen in picture (a) that NBSKP fibers are quite randomly dispersed in all directions. In picture (d) with closer look the connection between polymer and fiber is quite tight even though the polymer penetration into fiber network may be limited causing some voids inside the fiber structure (arrow in picture a), that is in relation to the lower density of the material. The picture (b) of the extrusion molded material shows how fibers are oriented to some extent in the flow direction, although clear agglomerates exist in the product. In extrusion the orientation of fibers into the melt flow direction is a typical behavior.[4] Picture (e), with greater enlargement for extrusion-formed material, shows that fibers are loosely connected to the polymer and they can be easily pulled out from the polymer matrix leaving holes in the polymer matrix.[11] In injection molded products, in pictures (c and f), the fibers are shorter and more randomly oriented than in the extrusion process. The fiber-polymer connection in picture (f) is tighter than in extrusion due to higher pressure packing the materials together during the injection molding process, that is, reflected also as a higher density presented in Table 1. Yet, the fibers can be easily torn off due to inadequate coupling of fibers and polymer matrix. The pictures reveal some porosity in materials, which is in relation to density values of materials presented in Table 1. The density values indicate that foam-formed materials contain more voids compared to extruded or injection molded materials and also that extruded material has slightly higher porosity than injection molded materials. This assumption can be verified from SEM pictures (a–c) in Figure 5.

Figure 6 shows fibers after processing, when the polymer matrix is dissolved away from 30% NBSKP fiber-

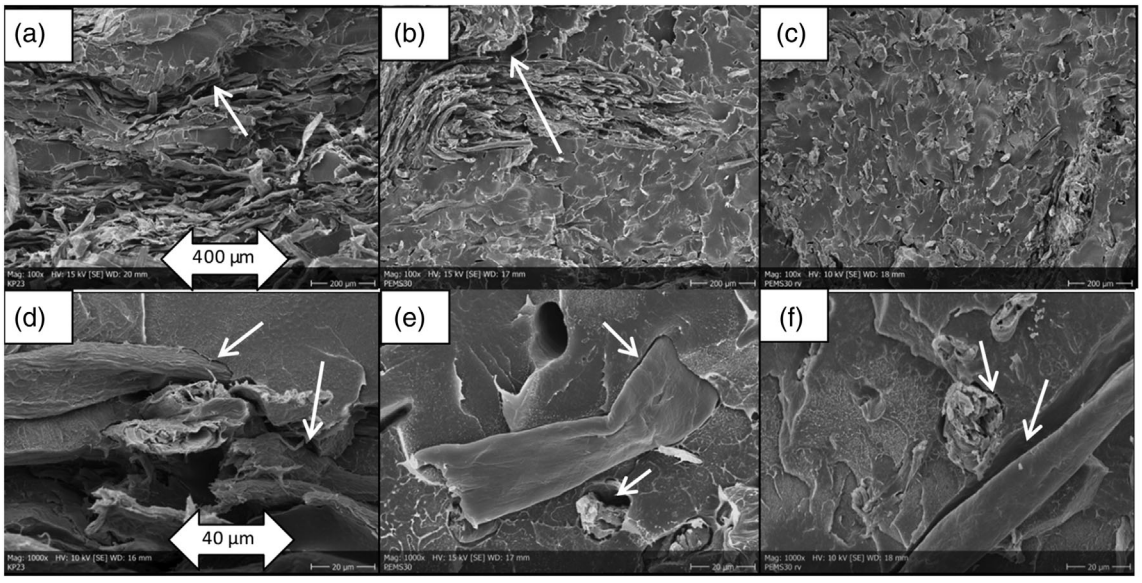

FIGURE 5 SEM pictures of composite materials containing 30% NBSKP. Pictures (a) and (d) are foam-formed material, (b and e) are extruded sheet, and (c and f) injection molded material. SEM pictures with ×100 enlargement are shown in the upper row and with ×1,000 enlargement in the bottom row. Arrows show some interphases between fibers and polymer and voids in the structure. NBSKP, northern bleached softwood kraft pulp

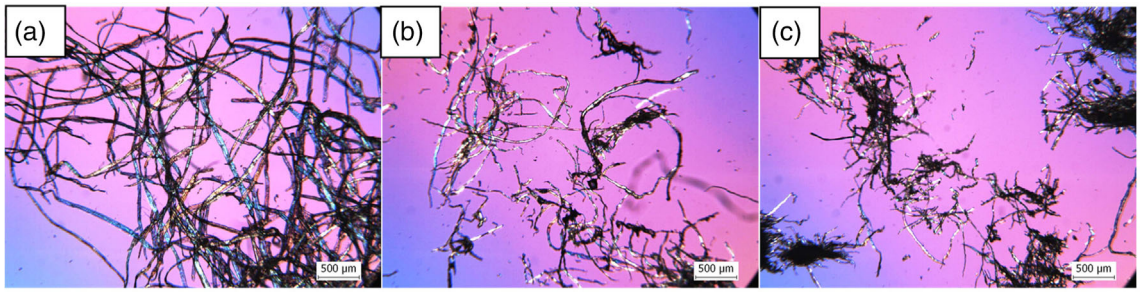

FIGURE 6 NBSKP fibers dissolved out from the products after processing. Picture (a) fibers from FF-NBSK30, (b) fibers from E-NBSK30, and (c) fibers from IM-NBSK30. NBSKP, northern bleached softwood kraft pulp [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 2 Results from particle size analysis for fibers separated from 30% fiber-containing composites

Sample code	Sample	Average particle size (µm)	90% of particles below (µm)
FF-NBSK30	Foam-formed NBSKP 30%, LDPE 70%	1,150	2,390
E-NBSK30	Extruded NBSKP 30%, LDPE 70%	82	429
IM-NBSK30	Injection molded NBSKP 30%, LDPE 70%	61	207

Abbreviation: NBSKP, northern bleached softwood kraft pulp.

TABLE 3 Results from tensile and Charpy impact strength tests

Sample code	Tensile strength (MPa)	Young modulus (MPa)	Strain at break (%)	Charpy impact strength (kJ/m^2)	Density (kg/m^3)
FF-NBSK100	1.06 ± 0.2	79 ± 23	3.4 ± 0.1	10.3 ± 1.4	0.41
FF-NBSK50	8.1 ± 0.5	706 ± 70	10.6 ± 2.7	64.6 ± 26	0.79
FF-NBSK30	11.9 ± 2	1,288 ± 240	7.3 ± 3.8	107 ± 29	0.93
E-NBSK30	8.3 ± 0.2	659 ± 252	8.0 ± 4.1	69.1 ± 28	1.002
IM-NBSK30	12.4 ± 0.3	543 ± 40	8.7 ± 0.6	35.5 ± 7.2	1.038
IM-NBSK50	13.6 ± 0.2	1,512 ± 104	2.0 ± 0.1	10.8 ± 1.3	1.144
IM-LDPE100	15.3 ± 0.1	142 ± 6	438 ± 37	No break (61.7 ± 1.5 as notched)	0.92

Abbreviation: NBSKP, northern bleached softwood kraft pulp.

containing samples. The results from the particle size analysis presented in Table 2 show the effect of more harsh process conditions for NBSKP fibers in extrusion and in injection molding compared to foam forming.

Figure 6a shows that fibers have retained their length in the foam-formed sample FF-NBSK30. Fibers in extruded sample E-NBSK30, in Figure 6b, and injection molded sample IM-NBSK30, in Figure 6c, are both cut and curved and the material contains smaller fiber fractions, which can be seen also in the particle size measurement results in Table 2. The amount of very small fiber fraction was the highest in the injection molded sample. The most challenging processing conditions occur in injection molding leading to the highest degradation of fibers. Similar cutting of fibers in thermoplastic composites due to the injection molding process is presented by Madsen et al.[24] and Shibata et al.[25]

3.2 | Mechanical properties

Mechanical properties of the materials were determined by measuring Charpy impact and tensile strength. The strength results are presented in Table 3 and Figures 7 and 8.

The tensile strength of the materials is presented in Table 3 and Figure 7a. The tensile strength is the highest in pure injection molded LDPE (15.3 MPa) and the

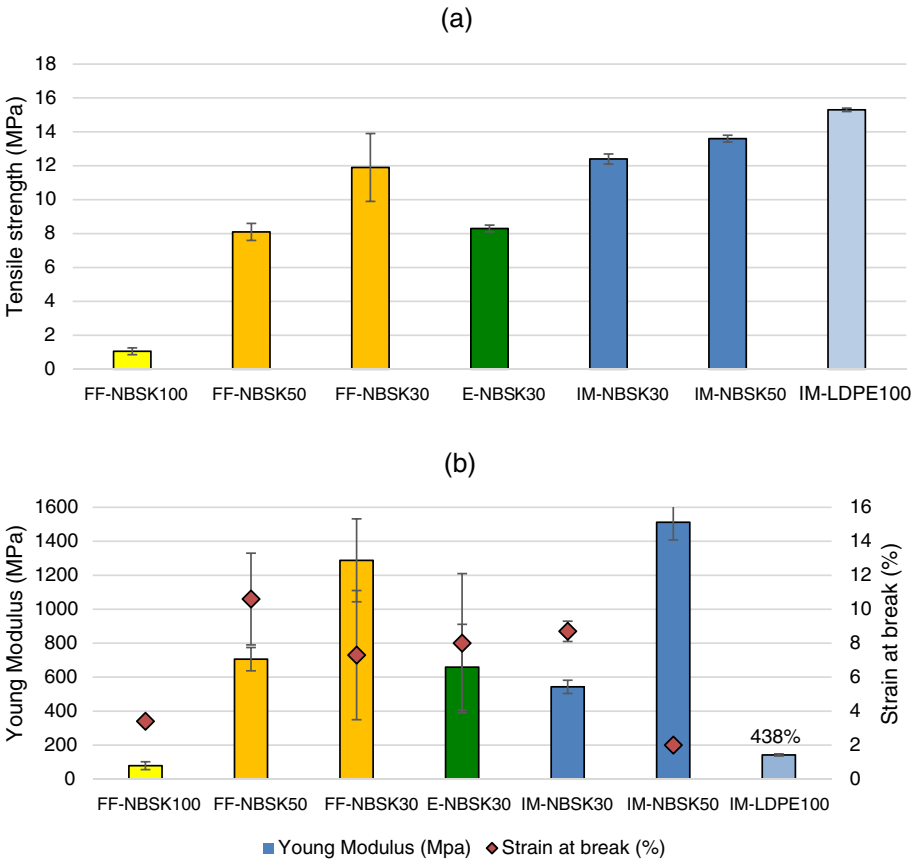

FIGURE 7 Tensile test results for foam-formed, extruded, and injection molded materials. (a) tensile strength results and (b) Young modulus and strain at break [Color figure can be viewed at wileyonlinelibrary.com]

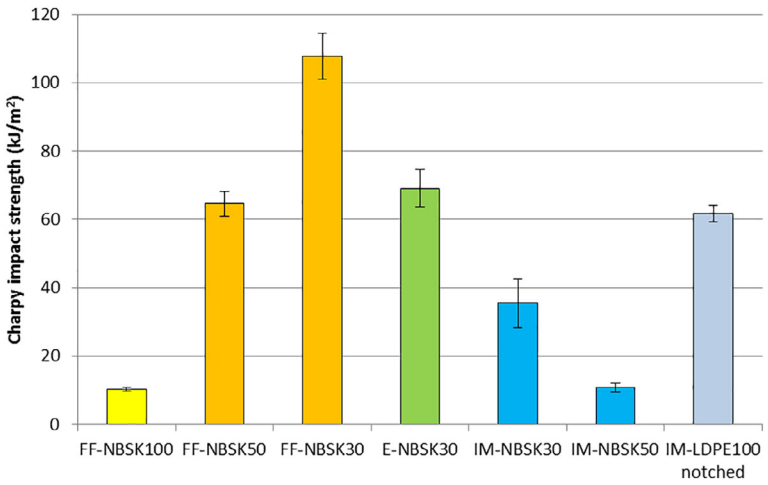

FIGURE 8 Charpy impact strength (un-notched) test results for foam-formed, extruded, and injection molded materials. IM-LDPE100 measured as notched due to no breakage as un-notched [Color figure can be viewed at wileyonlinelibrary.com]

lowest in foam-formed material (1.06 MPa) containing only NBSKP. Tensile strength of 30% fiber-containing injection molded material (12.4 MPa) have 4% higher tensile strength compared to 30% fiber-containing foam-formed material, and 33% higher compared to tensile strength value in extruded materials. This can be due to higher density, better fiber-polymer connection, and more homogeneous polymeric material in between short evenly distributed fibers compared to the foam-formed and extruded materials presented in Figure 5. The poor tensile strength in extruded material is partly due to fiber agglomerates presented in Figure 3 and poor fiber-polymer connection illustrated in SEM pictures (Figure 5b,e). In 50% fiber-containing materials the tensile strength in injection molded material (IM-NSBK50) (13.6 MPa) is 40% higher than in foam-formed material (FF-NSBK50), where LDPE does not form a uniform structure inside the fiber network, but more like clouds in between fibers, presented in Figure 4. The main tensile load in foam-formed material is transferred by a fiber

network and cut by polymer sections. When comparing the foam-formed materials FF-NSBK50 and FF-NSBK30, the material with lower fiber content has 32% better tensile strength (11.9 vs. 8.1 MPa). This is at least partly due to more continuous LDPE sections in foam-formed materials with a lower fiber content presented in Figure 4, and higher density (0.93 vs. 0.79 kg/m^3) close to neat LDPE (0.92 kg/m^3) indicating less porosity in the material with lower fiber content.

Tensile modulus is presented in Table 3 and Figure 7b. Neat LDPE and pure fiber network in FF-NSBK100 have quite low tensile modulus values of 142 and 49 MPa, respectively. In injection molded materials the tensile modulus increases with increasing fiber load, from 543 MPa in IM-NSBK30 to 1,512 MPa in IM-NSBK50. Similar phenomena are presented also by other authors with increasing filler contents.[26,27] In extrusion molded material with 30% fiber, the tensile modulus (659 MPa) is at the same level as injection molded material with the same fiber load considering the high standard deviation due to uneven material (E-NBSK30). Foam-formed material with higher fiber content (50%) has 45% lower tensile modulus (706 MPa) than 30% fiber-containing material (1,288 MPa), contrary to injection molded materials. The difference is due to lower density in FF-NSBK50 than FF-NSBK30, and the fact that the fiber network created during foam forming is controlling the load transfer in a material with 50% fiber load, causing the faster breakage of material during initial stretching of materials without any coupling agent. The foam-formed, 30% fiber-containing material (FF-NSBK30), however had 68% higher modulus than the corresponding injection molded material and also higher modulus than presented in the literature for similar injection molded material.[28,29] This improvement comes through longer fibers and fiber network enabling load transfer.

The strain at break presented in Table 3 and Figure 7b collapses as soon as fibers are introduced in LDPE, which is logical due to increased fraction points in the material caused by poor fiber-polymer interaction. In all materials with 30% fiber content, the elongation is 11–13% of neat LDPE (438%) due to a dominating polymer fraction. In injection molded material with 50% fibers (IM-NSBK50), containing also very short fiber fractions and poor fiber-polymer connection, elongation is only 2% due to a higher amount of potential fraction points. In foam-formed material with 50% fiber (FF-NSBK50) the elongation is 10.6%. In FF-NSBK50 the long unbroken fibers provide less fraction points than in injection molded material with 50% fiber. FF-NSBK100 shows the elongation for pure fiber network as 3.4%.

The high impact strength in foam-formed composite materials presented in Figure 8 and Table 3 is due to long intact fibers combined with a continuous polymer matrix inside the composite structure illustrated in Figures 5a and 6a. Especially in FF-NSBK30, the partly continuous polymer sections are reinforced with fiber networks providing high impact strength (107 kJ/m^2). The very short fibers spread randomly in injection molded material, presented in Figures 5c and 6c, act like breaking points during the impact strength test for both materials, with 30 and 50% fibers. This phenomena can be seen especially with higher fiber content material, IM-NSBK50, and reflected as low impact strength value 10.8 kJ/m^2, which result is at the same level as in commercial LDPE composites and previously presented in other researches.[23,28,29] The impact strength in injection molded 30% NBSKP fiber-containing LDPE composite was 35.5 kJ/m^2, being only 33% of comparative foam-formed composite impact strength. In extruded material (E-NSBK30) there are more continuous polymer parts due to high agglomeration of fibers, but also longer fibers than in injection molded material giving higher impact strength than injection molded material.

3.3 | Economic evaluation

The biggest benefit of the foam forming process compared to extrusion or injection molding is that fiber length is maintained during the processing of composite materials. Long intact fibers in composites enable the achievement of high impact strength. Impact behavior is an important mechanical property of engineering materials used for many popular applications including interior and exterior components of automobiles, buildings, and aircrafts.[30,31]

The economic evaluation is based on three trial points E-NBSK30, IM-NBSK30, and FF-NBSK50, because the mechanical strength properties and especially Charpy impact strength is at the same level in E-NBSK30 and FF-NBSK50 (Figure 8), and in IM-NBSK30 the proportion of softwood fibers is the same as in E-NBSK30. Here it should be noted that the calculation is made in metric tons and that in injection molding end products are mostly very different than in extrusion. Here the position of foam forming technology, from a cost point of view, for both extrusion and injection molding technologies is shown.

Capacities and operation hours are assumed to be following: Extrusion (E-NBSK30): 2,000 t/a,[32] injection molding (IM-NBSK30) 1,000 t/a,[32] and foam forming (FF-NBSK50) 30,000 t/a.[33] Annual hours of operation are estimated as 5,280 hr. Planned maintenance breaks

are considered to be made outside operation hours (during weekends and holidays).

Personnel is considered to work 8 hr per shift for five working days per week, for 11 months, having 20 operations days per month in three shifts during operation. This results in a sum of 5,280 working hours. Modern extrusion and injection molding lines can operate with two persons per shift. For the foam forming line, the needed personnel per shift is assumed to be five.

For labor, the cost per hour is assumed to be 25 €/hr, including overhead of about 5 €/hr, which is the average hourly wages in the EU-28. For extrusion, with operating hours of 5,280 hr/a (person hours 10,560 hr/a), direct personnel costs are annually 211,200 €/a, and additional personnel costs add 25% (52,800 €/a), giving total personnel cost of 144 €/t. For injection molding, similar personnel costs are assumed, but with half of the capacity of the line. For injection molding, the personnel cost with a similar approach is 263 €/t. For foam forming, personnel cost is 22 €/t, with total personnel hours of 26,400 hr/a.

Cost of energy is assumed to be the European average for a user of this scale and utilization of 1.5 and 3.2 kWh/kg[34] for extrusion and injection molding respectively for the site. Extrusion extruder consumes 50%, chiller 26%, compressed air 11%, and water pumps 5% of the overall site consumption. Injection molding main process consumes 62.5%, chiller and cooling water 15.2%, compressed air 9%, and water pumps 8.1% of the overall site consumption.

With electrical energy as the primary source, the cost is 118 and 252 €/t for extrusion and injection molding, respectively, and 18 €/t for foam forming. For foam forming the energy requirement for drying associated with this production is annually ~0.79 MWhr/t_{water}. Drying is assumed to begin at a moisture ratio of 0.67 (dry solid content 60%) and accomplished using impingement and through air drying, summing to 18 €/t. Energy values in Table 4 are typical prices for an EU-28 industrial customer of this scale without taxes during the second half of 2015.

Water consumption is assumed for foam forming. For foam forming the use of water is assumed to be 10 m^3/t[38] (9 €/t) with assumed water cost of 0.85 €/m^3 (Pretreatment[39] and Wastewater[40]) and partial recovery of water (as the 2.3% consistency without recovery would use 42.5 m^3). For injection molding and extrusion cooling water circulation is closed and thus omitted.

Manufacturing expenses, assumed from an *investment* of 0.8 M€ for an extrusion of this size[32,41,42] and 0.2 M€ for injection molding.[32,43,44] With a depreciation period of 10 years, the annual depreciation cost without capital cost is thus 39 and 19 €/t, respectively. Mold depreciation in injection molding is assumed to be identical and equal to machine depreciation, 19 €/t.[32] However, this may vary from application to application.[43] Here the potential is calculated from the plasticizing capacity, but in practice the OEE (combined planned availability, quality, and performance) can vary from 30 to 90%[45–49] and in injection molding even 20%.[50] For comparison, OEE of Foam line is expected to be in the upper limit, 70–80%.[51]

Foam line investment is estimated to be 7 M€ (investment cost of 220 €/t, 10 year depreciation time gives annually 22 €/t).[52]

Direct *material costs* are calculated from the raw material prices. Prices of raw materials are assumed to be the following: NBSK 541 €/t, LDPE 1,365 €/t, PE-MAH 1,280 €/t, and TPW 9,800 €/t.[53] Cost of SDS is assumed to be 1,500 €/t[54] and included in raw material cost.

This results in a sum of 1,205 €/t for raw material prices for extrusion and injection molding and 1,043 €/t for foam forming.

TABLE 4 Main assumptions

Item	Value
Personnel	20 €/hr (+25% other costs)[35]
Energy	Natural gas for heat 25.6 €/MWhr[36] and electricity 78.8 €/MWhr[37]
Water	0.85 €/m^3

TABLE 5 Processing cost (€/t) at the site with the compared cases for manufacturing one ton of end products

Cost	E-NBSK30 (€/t)	IM-NBSK30 (€/t)	FF-NBSK50 (€/t)
Personnel	144	263	22
Energy	118	252	18
Water			9
Raw materials	1,205	1,205	1,043
Investment	39	38	22
Total	1,506	1,758	1,114

Abbreviation: NBSKP, northern bleached softwood kraft pulp.

Summary of processing costs at the site (without engineering, administrative, and marketing costs and contingencies and profit) are thus 1,506 €/t for extrusion case, 1,758 €/t for injection case, and 1,114 €/t for foam forming case (see Table 5). The share presented is similar to the estimate presented by Euromap.[53]

With the assumptions used, the total production costs at the site are lowest with the foam forming-case and highest with the injection molding-case.

4 | CONCLUSIONS

As conclusions, it can be said that when compared to cellulose fiber-containing polyethylene composites manufactured by extrusion or injection molding, foam forming seems to be, from a strength perspective, a wise and feasible production method for manufacturing sheet-form thermoplastic materials, when the fiber content is below 50%. In applications where the material impact properties, stiffness, and visual appearance are the most significant measures, the foam forming process provides composite materials where fiber degradation is minimized due to low temperatures and less or no shearing forces breaking the fiber during processing. This enabled three times higher Charpy impact strength properties for 30% NBSKP fiber-containing LDPE composite prepared by foam-formed material compared to injection molding. The tensile modulus of injection molded 30% NBSKP fiber-containing material was 68% lower than similar material prepared by foam forming. Also, the color of the foam-formed composite stays light, almost without color formation due fiber degradation. It is also a potential production method for high fiber content thermoplastic composites with fiber content over 50%, if uniform-quality fiber dispersion can be provided with the help of dispersing additives introduced in one process during foam formation.

The cost reduction of foam forming against extrusion or injection molding is found to be between 26 and 37%. This arises mainly from lower personnel costs, raw material prices, and energy costs. Foam forming has a low need for preprocessing treatments of cellulose fibers, for example, pelletizing and drying processes normally used for fibers before plastic processing, which makes foam forming a potential manufacturing method for thermoplastic polymer-cellulose fiber composites. Foam forming enables larger production, like the commercial reference shows for range, compared to the other two technologies and thus it is suitable for manufacturing products with high production volumes, which also supports savings potential.

The results show that this kind of technology has a promising potential in numerous applications utilizing thermoforming through pressing in industrial fields and in consumers' daily lives.

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